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Dimensional crossover field as a function of oxygen stoichiometry in $YBa_2Cu_3O_{7-\delta}$ thin films

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The magnetic phase diagram of $YBa_2Cu_3O_{7-\delta}$ thin films was studied as a function of anisotropy by systematically changing the oxygen stoichiometry (δ value). For small δ values the vortex-solid phase transition boundary $H_g(T)$ is observed over the full accessible field range (50 kOe) to follow the $(1-T/T_c)^{\sim 1.5}$ dependence that is expected for an anisotropic superconductor in the three-dimensional regime. As δ increases, a portion of the phase boundary no longer follows the $(1-T/T_c)^{\sim 1.5}$ dependence making it possible to identify the anisotropy-dependent dimensional crossover field H_0 at which the system begins to behave quasi-two-dimensionally. Plotting $H_g(T)$ normalized by H_0 as a function of $(1-T/T_c)$ results in a collapse of the phase boundaries onto a single universal curve. Using published values of the anisotropy $\gamma=8.5$ and the upper critical field slope $dH_{c2}/dT=-20$ kOe/K it is possible to determine the specific relation $H_0 \approx 1.2\phi_0/(d^2\gamma^2)$, where d is the multilayer spacing, which can be used to determine the δ dependence of γ.

The CuO-based high-temperature superconductors (HTS's) represent a group of highly anisotropic materials that are often described as alternating layers of superconducting and normal, or insulating, layers.[1] Numerous experiments have suggested that superconductivity resides primarily in a layer composed of nearly planar CuO_2 multilayers with the alternate layer functioning as charge-reservoir layer that affects the carrier concentration in the CuO_2 multilayers.[2] Flux vortices in these materials are often described as supercurrent loops within the CuO_2 multilayers (pancake vortices) strung together into flexible vortex lines.[3] The pancake vortices in different CuO_2 multilayers can be coupled via their magnetic interactions or Josephson coupling, with the strength of this coupling depending on the anisotropy of the material.[1]

Several theoretical models have been proposed to describe the flux-vortex behavior and its implications for the magnetic phase diagram of anisotropic superconductors like the HTS's.[4–6] For the more weakly anisotropic HTS's (e.g., $YBa_2Cu_3O_7$) the model of Feigel'man, Geshkenbein, and Larkin (FGL),[5] as well as that of Fisher, Fisher, and Huse (FFH),[6] describe a phase transition below the upper critical field H_{c2} at which point a liquid of flexible flux vortices becomes a flux-vortex solid (glass or crystal). In the case of strongly disordered materials (e.g., $YBa_2Cu_3O_7$ thin films) a continuous vortex-glass phase transition is expected and has been observed.[7] At a sufficiently large applied magnetic field value, the system will behave quasi-two-dimensionally as a result of fluctuations rendering the pancake vortices within a given CuO_2 multilayer independent of the pancake vortices in neighboring CuO_2 multilayers.[4–6] The value of this dimensional-crossover field H_0 is expected to be proportional to $\phi_0/(d\gamma)^2$, decreasing with the anisotropy $\gamma=\xi_{ab}/\xi_c$; where d is the distance between CuO_2 multilayers and the coherence lengths ξ_{ab} and ξ_c are in the ab plane and along the c axis, respectively.[4–6] Here we present a specific determination of H_0 and show how it evolves with changes in anisotropy that are produced in epitaxial thin films of $YBa_2Cu_3O_{7-\delta}$ (YBCO) as oxygen is systematically removed.

Epitaxial YBCO thin films, with the c axis perpendicular to the film plane, were prepared by pulsed laser deposition on heated (001) $LaAlO_3$ substrates as previously described.[8] Each thin-film sample was about 4000 Å thick and initially had a superconducting transition temperature $T_c=90\pm2$ K, with a transition width ΔT_c of about 2 K when measured using ac susceptibility (χ_{ac}) at 1.5 MHz. The oxygen stoichiometry of these films was modified by annealing at 400 °C in a controlled flowing mixture of oxygen and argon gases followed by a rapid quench to room temperature as described previously.[9] A separate film was prepared for each oxygen stoichiometry. The T_c value for a given δ value correspond closely with previous studies[10] and there is an estimated error of about 0.05 in the values of δ used here.

The films were patterned using laser ablation into bridges having dimensions of 100 μm wide by 2000 μm long. A standard four-point probe technique was used for the temperature-dependent resistivity measurements $\rho(T)$ and electric field as a function of current density $E(J)$ measurements using procedures described previously.[11] The dc magnetic field (H), ranging from 0 to 50 kOe, was applied parallel to the crystallographic c axis.

Shown in Fig. 1 are the vortex-glass phase boundaries $H_g(T)$ for δ ranging from 0.05 to 0.65. The points at 10 and 50 kOe were determined by scaling of $E(J)$ curves, while those at the other fields studied were determined by analyzing $\rho(T)$ using a linear extrapolation to the temperature at which the quantity $[d(\ln\rho)/dT]^{-1}$ equals zero.[12–15] The $H=0$ kOe points are the T_c values determined using $\rho(T)$ and χ_{ac} measurements. The $H_g(T)$ curves broaden significantly as δ increases, with the temperature difference $\Delta T=(T_c-T_g)$ at $H=50$ kOe equal to about 10 K for $\delta=0.05$ increasing to $\Delta T\sim35$ K for $\delta=0.65$; where the T_c value at $H=0$ is used. The temperature dependence of H_g is observed to change with δ. For $\delta<0.12$, the relation $H_g(T)\sim(1-T/T_c)^x$ with $x=1.3-1.5$ fits the results, which is consistent with previous observations of $H_g(T)$ in anisotropic three-dimensional high-temperature superconductors.[13] In contrast to this, for $\delta>0.12$, $H_g(T)$ can no

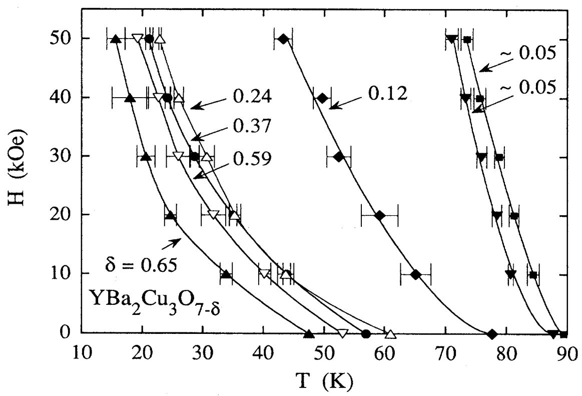

FIG. 1. Vortex-glass phase boundaries $H_g(T)$ determined for $YBa_2Cu_3O_{7-\delta}$ thin films with values of δ ranging from 0.05 to 0.65 using a vortex-glass scaling analysis of the electric field as a function of current density data at 10 and 50 kOe and the resistivity as a function of temperature data at all other fields.

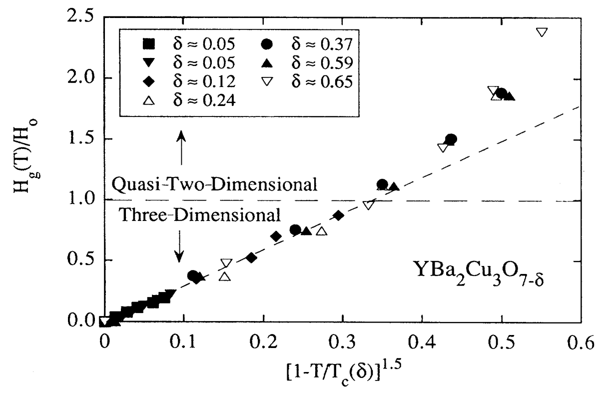

FIG. 2. The $H_g(T)$ boundaries shown in Fig. 1 normalized by the dimensional crossover field H_0 and plotted against $[1-T/T_c(\delta)]^{1.5}$. The universal $H_g(T)$ boundary in this plot deviates from linearity at the crossover between three-dimensional and quasi-two-dimensional behavior, which is found to occur at $H_g(T)/H_0=1$. This normalization procedure allows the determination of H_0 and the relative values of anisotropy $\gamma=\xi_{ab}/\xi_c$ for all δ values studied.

longer be described by a simple power-law temperature dependence over the full H range. The behavior in this regime is more consistent with observations of the vortex-glass transition in quasi-two-dimensional superconductors, such as $Tl_2Ba_2CaCu_2O_8$ (Ref. 14) and $Bi_2Sr_2CaCu_2O_x$ (Ref. 15), where at high field $H_g(T)$ decreases more rapidly with T.

The observed shift of the $H_g(T)$ phase boundary toward lower temperature is expected when the anisotropy increases.[9] A previous analysis of torque magnetometry data on grain-aligned YBCO powder samples[16] has shown γ to increase from about 7 to 32 as δ is increased from 0 to 0.5. These results are similar to the results described below. The increase in δ also corresponds to a decrease in the zero-temperature c-axis coherence length $\xi_c(0)$, which should result in a decrease of the Josephson coupling.[9] Either effect is consistent with YBCO moving closer to the two-dimensional limit as δ is increased. The FGL and FFH models both describe a dimensional crossover field that depends on the anisotropy γ as $H_0 \propto \phi_0/(\gamma d)^2$ (Ref. 5). If the δ dependence of $H_g(T)$ is a direct result of the δ dependence of H_0, then a plot of $H_g(T)/H_0$ as a function of $(1-T/T_c)$, where T_c is the δ-dependent superconducting transition temperature determined for each film, may be universal. By plotting $H_g(T)/H_0$ as a function of $(1-T/T_c)^{1.5}$ the three-dimensional portion of the H-T phase diagram that is below H_0 should be linear while the portion of the phase diagram above H_0, where $H_g(T)$ no longer shows this power-law temperature dependence, should deviate from linearity.

Figure 2 shows normalization of the $H_g(T)$ curves in Fig. 1, with a break from linearity at $H_g(T)/H_0 \approx 1$. Data collapse was accomplished by first determining the anisotropy value $\gamma=8.5$ for the film with $\delta=0.05\pm0.05$ ($T_c=89.4$ K). This analysis was originally attempted using γ in the range of 5–6 previously reported for stoichiometric YBCO crystals and powders,[16] however, this range of γ values was found to yield unreasonably large values for dH_{c2}/dT. Considering the fact that stoichiometric YBCO typically has a $T_c \approx 92$ K, it is likely that the 89.4 K thin film is not at the optimal oxygen content, and is thus likely to have a greater anisotropy value. The anisotropy value for this film was thus determined using the relation $\gamma=\{\phi_0/[2\pi\xi_c(0)^2 T_c|dH_{c2}/dT|]\}^{1/2}$ along with the upper critical field slope $dH_{c2}/dT=-20$ kOe/K determined for a YBCO thin film with $\delta=0.05$ (Ref. 17) and the zero-temperature c-axis coherence length $\xi_c(0)$ previously determined for this film to be 1.6 Å (Ref. 9) using the crossover temperature T_{23} from two- to three-dimensional fluctuations above T_c and the relation $\xi_c(0)=d/2(T_{23}/T_c-1)^{1/2}$ (Ref. 18). The quantity $H_g(T)/H_0$ was then determined for each of the other YBCO thin films by adjusting the γ value for each of these $H_g(T)/H_0$ curves in order that these curves collapsed onto the universal $H_g(T)/H_0$ versus $(1-T/T_c)^{1.5}$ curve determined for the film with $T_c=89.4$ K. The value of d for all films was fixed at 11.7 Å since x-ray-diffraction measurements on these films indicate that d varies from 11.70 to 11.77 Å as δ changes from 0.05 to 0.65. It was then found that this universal $H_g(T)/H_0$ versus $(1-T/T_c)^{1.5}$ curve deviates from linearity (no longer shows three-dimensional behavior) at $H_g(T)/H_0=1$, if $H_0 = 1.2\phi_0/(\gamma d)^2$. This relation is consistent with the models of FFH and FGL, which give order of magnitude estimates for H_0 of $\phi_0/(d\gamma)^2$ and $4\phi_0/(d\gamma)^2$, respectively, and it may apply to all high-temperature superconductors.[5,6] The $H_g(T)$ phase boundary determined using $\rho(T)$ data for a $Tl_2Ba_2CaCu_2O_8$ (Tl-2212) thin film was scaled onto the $H_g(T)/H_0$ curve in Fig. 2 with a value of $\gamma=42$ by first fixing $d=14.7$ Å (the c-axis length divided by the number of multilayers per unit cell) and then varying γ. This is in agreement with previous observations for Tl-2212 which show anisotropy values ranging from about 5 to 70.[19]

The values of the anisotropy γ and the dimensional crossover field H_0 determined using this scaling analysis are shown as a function of δ in Fig. 3. The γ values are consistent with previous measurements. For example, the value $\gamma=30$ for the $\delta=0.65$ sample is close to the previously reported value of $\gamma \approx 32$ determined from an analysis of torque magnetometry measurements on the grain-aligned YBCO

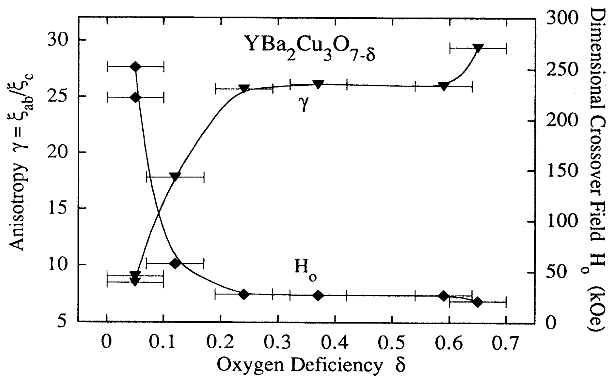

FIG. 3. The anisotropy γ and dimensional crossover field H_0 values, determined using the normalization procedure shown in Fig. 2, plotted as a function of δ for several oxygen-deficient $YBa_2Cu_3O_{7-\delta}$ thin films.

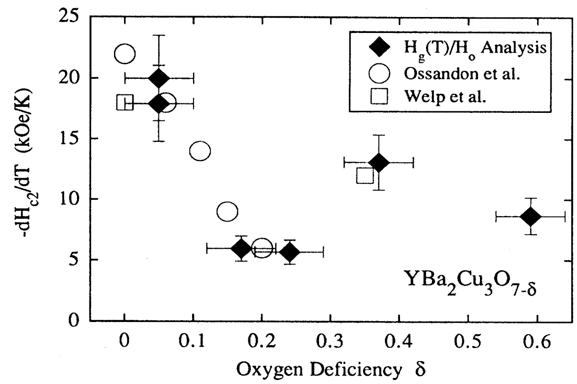

FIG. 4. The slope at T_c of the upper critical field as a function of δ. Solid diamonds represent values determined using $-dH_{c2}/dT=\phi_0[2\pi\gamma^2\xi_c(0)^2T_c]$, with $\xi_c(0)$ determined from the zero-field fluctuation conductivity at $T>T_c$ and γ values determined from the $H_g(T)/H_0$ analysis. Hollow circles are values determined by Ossandon *et al.* (Ref. 17) using a three-dimensional scaling analysis for $\delta=0$ and the hollow squares are values determined by Welp *et al.* (Ref. 20) using a two-dimensional scaling analysis for $\delta=0.35$.

powder sample with $\delta\approx0.5$ (Ref. 16). Note, however, that in previous analyses of γ as a function of δ on grain-aligned YBCO powder samples, γ increases slowly for $\delta=0$–0.25 (Ref. 16). In contrast to this, the results in Fig. 3 indicate that for thin films prepared under the conditions utilized here, γ increases quickly in the range $\delta=0$–0.25 and then plateaus in the range $\delta=0.25$–0.35 coincident with the 60 K T_c plateau. The $H_0(\delta)$ plot in Fig. 3 also shows a rapid decrease with δ in the range $\delta=0$–0.25, dropping from about 260 kOe to about 30 kOe. The small values determined for H_0 when $\delta>0.25$ are consistent with the work of Welp *et al.*, which found that the $M(T)$ isochamps measured from $H=10$ to 50 kOe for YBCO with $\delta\approx0.35$ are better described by a two-dimensional, rather than a three-dimensional, fluctuation scaling analysis.[20]

Shown in Fig. 4 are the values of dH_{c2}/dT plotted as a function of δ for YBCO thin films with H parallel to the c axis using the Ginzburg-Landau relation $-dH_{c2}/dT=\phi_0/[2\pi\xi_{ab}^2(0)T_c]$ (Ref. 20) and the definition of anisotropy $\gamma=\xi_{ab}/\xi_c$. The values of dH_{c2}/dT represented by the solid diamonds were determined using the γ values plotted in Fig. 3 and the $\xi_c(0)$ values determined for the same films using a zero-magnetic-field fluctuation conductivity analysis.[9] The hollow circles represent dH_{c2}/dT values determined by Ossandon *et al.* for $\delta<0.2$ using the Hao-Clem magnetization analysis, which is based on the anisotropic Ginburg-Landau theory.[17,21] These different determinations of dH_{c2}/dT indicate that the magnitude of dH_{c2} decreases as δ increases to 0.2. However, the present analysis extends the range over which dH_{c2}/dT has been determined and indicates that dH_{c2}/dT exhibits a minimum between the 90 and 60 K plateaus. These results are in agreement with both the two-dimensional scaling analysis of Welp *et al.* (Ref. 20) at large δ values ($dH_{c2}/dT\approx-12$ kOe/K for $\delta\approx0.35$) as well as the values of dH_{c2}/dT determined at small δ ($\delta<0.2$) by Ossandon *et al.*[17]

In summary, the temperature dependence of the vortex glass transition $[H_g(T)]$ was studied in thin films of $YBa_2Cu_3O_{7-\delta}$ as a function of δ. The dimensional crossover field H_0 was identified using the deviation in the temperature dependence from $H_g\sim(1-T/T_c)^{1.5}$ expected in the three-dimensional regime. The $H_g(T)$ curves at all δ values could be collapsed onto one another by appropriate normalization of both the T scale and the H scale. Using the relation $H_0\sim\phi_0/(\gamma d)^2$ with values of γ and d determined for nearly fully oxygenated YBCO thin films, it was possible to determine relative values of the anisotropy γ as a function of δ. There is a notable generality of the phase diagram and an attempt to collapse the $H_g(T)$ curve of Tl-2212 onto the YBCO results met with success. It is possible that the specific relation $H_0\approx1.2\phi_0/(\gamma d)^2$ determined may be generally applicable to CuO-based HTS materials, thereby providing a general method for determining the anisotropy in HTS materials from transport data.

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[1] J. R. Clem and M. W. Coffey, Phys. Rev. B **42**, 6209 (1990); J. R. Clem, *ibid.* **43**, 7837 (1991).

[2] R. J. Cava, Science **247**, 656 (1990).

[3] S. Doniach, in *The Phenomenology of Flux Motion in High Temperature Superconductors*, Proceedings of the Los Alamos Symposium—1989, edited by K. S. Bedell, D. Coffey, D. E. Meltzer, D. Pines, and J. R. Schrieffer (Addison-Wesley, Reading, MA, 1989), p. 406.

[4] L. I. Glazman and A. E. Koshelev, Phys. Rev. B **43**, 2835 (1991).

[5] M. V. Feigel'man, V. B. Geshkenbein, and A. I. Larkin, Physica C **167**, 177 (1990).

[6] M. P. A. Fisher, Phys. Rev. Lett. **62**, 1415 (1989); D. S. Fisher, M.

P. A. Fisher, and D. A. Huse, Phys. Rev. B **43**, 130 (1991).

[7] R. H. Koch, V. Foglietti, W. J. Gallagher, G. Koren, A. Gupta, and M. P. A. Fisher, Phys. Rev. Lett. **64**, 2586 (1990).

[8] R. E. Muenchausen, K. M. Hubbard, S. R. Foltyn, R. C. Estler, and N. S. Nogar, Appl. Phys. Lett. **56**, 578 (1990); X. D. Wu, R. E. Muenchausen, S. R. Foltyn, R. C. Estler, R. C. Dye, C. Flamme, N. S. Nogar, A. R. Garcia, J. Martin, and J. Tesmen, *ibid.* **56**, 1481 (1990).

[9] L. Hou, J. Deak, P. Metcalf, and M. McElfresh, Phys. Rev. B **50**, 7226 (1994).

[10] R. J. Cava, A. W. Hewat, E. A. Hewat, B. Batlogg, M. Marezio, K. M. Rabe, J. J. Krajewski, W. F. Peck, Jr., and L. W. Rupp, Jr., Physica C **165**, 419 (1990); E. Osquiguil, M. Maenhoudt, B. Wuyts, and Y. Bruynseraede, Appl. Phys. Lett. **60**, 1627 (1992); E. C. Jones, D. K. Christen, J. R. Thompson, R. Feenstra, S. Zhu, D. H. Lowndes, J. M. Phillips, M. P. Siegal, and J. D. Budai, Phys. Rev. B **47**, 8986 (1993).

[11] J. Deak, M. McElfresh, J. R. Clem, Z. Hao, M. Konczykowski, R. Muenchausen, S. Foltyn, and R. Dye, Phys. Rev. B **47**, 8377 (1993).

[12] J. Deak and M. McElfresh, Phys. Rev. B **48**, 1337 (1993).

[13] P. L. Gammel, L. F. Schneemeyer, and D. J. Bishop, Phys. Rev. Lett. **66**, 953 (1991).

[14] J. Deak, M. McElfresh, D. W. Face, and W. L. Holstein (unpublished).

[15] H. Safar, P. L. Gammel, D. J. Bishop, D. B. Mitzi, and A. Kapitulnik, Phys. Rev. Lett. **68**, 2672 (1992).

[16] B. Janossy, D. Prost, S. Pekker, and L. Fruchter, Physica C **181**, 51 (1991).

[17] J. G. Ossandon, J. R. Thompson, D. K. Cristen, B. C. Sales, H. R. Kerchner, J. O. Thompson, Y. R. Sun, K. W. Lay, and J. E. Tkaczyk, Phys. Rev. B **45**, 12 534 (1992); E. C. Jones, D. K. Christen, J. R. Thompson, J. G. Ossandon, R. Feenstra, Julia M. Philips, and M. P. Siegal, *ibid.* **49**, 572 (1994).

[18] B. Oh, K. Char, A. D. Kent, M. Naito, M. R. Beasley, T. H. Geballe, R. H. Hammond, A. Kapitulnik, and J. M. Graybeal, Phys. Rev. B **37**, 7861 (1988).

[19] J. H. Kang, K. E. Gray, R. T. Kampwirth, and D. W. Day, Appl. Phys. Lett. **53**, 2560 (1988); H. Mukaida, K. Kawaguchi, M. Nakao, H. Kumakura, D. R. Dietderich, and K. Togano, Phys. Rev. B **42**, 2659 (1990).

[20] U. Welp, S. Fleshler, W. K. Kwok, R. A. Klemm, V. M. Vinokur, J. Downey, and G. W. Crabtree, *Reversible Magnetization of Copper Oxide Superconductors in the Mixed State*, in *High Temperature Superconductivity*, edited by S. K. Malik and S. S. Shah (Nova Science, New York, 1992).

[21] Z. Hao, J. R. Clem, M. McElfresh, L. Civale, A. P. Malozemoff, and F. Holtzberg, Phys. Rev. B **43**, 2845 (1991).